

Assessing the spatial and temporal variability of fine particulate matter components in Israeli, Jordanian, and Palestinian cities

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ABSTRACT

This manuscript presents results from an extensive, multi-country comparative monitoring study of fine particulate matter (PM_{2.5}) and its primary chemical components in Israeli, Jordanian and Palestinian cities. This study represented the first time that researchers from these countries have worked together to examine spatial and temporal relationships for PM_{2.5} and its major components among the study sites. The findings indicated that total PM_{2.5} mass was relatively homogenous among many of the 11 sites as shown from strong between-site correlations. Mean annual concentrations ranged from 19.9 to 34.9 $\mu\text{g m}^{-3}$ in Haifa and Amman, respectively, and exceeded accepted international air quality standards for annual PM_{2.5} mass. Similarity of total mass was largely driven by SO_4^{2-} and crustal PM_{2.5} components. Despite the close proximity of the seven, well correlated sites with respect to PM_{2.5}, there were pronounced differences among the cities for EC and, to a lesser degree, OC. EC, in particular, exhibited spatiotemporal trends that were indicative of strong local source contributions. Interestingly, there were moderate to strong EC correlations ($r > 0.65$) among the large metropolitan cities, West Jerusalem, Tel Aviv and Amman. For these relatively large cities, (i.e., West Jerusalem, Tel Aviv and Amman), EC sources from the fleet of buses and cars typical for many urban areas predominate and likely drive spatiotemporal EC distributions. As new airshed management strategies and public health interventions are implemented throughout the Middle East, our findings support regulatory strategies that target integrated regional and local control strategies to reduce PM_{2.5} mass and specific components suspected to drive adverse health effects of particulate matter exposure.

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1. Introduction

In the Middle East and North Africa, urban air pollution has been identified as a leading factor contributing to the regional burden of disease (World Health Organization, 2001). For many countries of this region, however, estimating population exposures to urban air pollution and true health risk, is difficult. Information concerning air pollutant levels, trends and transboundary impacts is virtually non-existent in much of the Middle East. Previous studies of aerosol fate and transport in this region have highlighted the impact of several long-range transport processes to sites primarily within Israel. Typically, these studies focus on transport of crustal aerosol from North Africa and anthropogenic particle mass from Europe (Luria et al., 1986, 1996; Graham et al., 2004; Derimian et al., 2007; Asaf et al., 2008; Mamane et al., 2008; Weinroth et al., 2008). Several

studies have also examined wet and dry deposition processes for the removal of anthropogenic and natural sources of urban pollution in Jordan (Al-Momani et al., 2002, 2008).

Similar to the U.S. and Europe, there is debate in this region about the most effective approaches for reducing levels of ambient particulate matter (PM) and PM-associated health impacts. While there have been few studies examining air pollution-related exposures or health effects in the Middle East (Braunstein and Goren, 2000; Hellman and Goren, 2002; Kordysh et al., 2005), it is likely that PM pollution is responsible for excess morbidity and mortality in this region as well (Le Tertre et al., 2005).

Effective regulatory control of ambient PM in the Middle East is complicated by geopolitical tensions, which have limited integrated, regional management strategies, as well as scientific uncertainty regarding the transboundary impacts of PM emission and sources among the countries of the region. Given the close geographic proximity of many Middle Eastern cities to each other, the influence of shared meteorology and lack of substantial geophysical barriers separating cities of the region, it is reasonable to assume that emission impacts and/or airshed management practices in one

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locale may affect PM concentrations regionally. To date, however, little is known about the relative contribution of regional and local sources in Middle Eastern cities and how the concentrations of PM components in these cities correlate with each other. A better understanding of PM composition and correlations among Middle Eastern cities is a necessary first step in determining the regional vs. local contributions to this pollutant, developing effective pollution control strategies, and ultimately, reducing the public health burden from PM-associated morbidity and mortality.

To investigate seasonal trends and correlations in PM among cities in the Middle East, we conducted an extensive, multi-country comparative monitoring study of fine particulate matter ($PM_{2.5}$) and its primary chemical components in Israeli, Jordanian and Palestinian cities. This study is among the first efforts of researchers from these countries to jointly examine PM air pollution in a scientific setting. While a complete investigation of between-site and transboundary $PM_{2.5}$ impacts is beyond the scope of this study, the current analysis provides initial information concerning the spatial and temporal relationships for $PM_{2.5}$ and its major components among the study sites. Together, these findings provide insight into shared sources and the relative impact of both regional and local PM to direct future examinations of PM regional transport and potential strategies for the effective control of this pollutant.

2. Methods

$PM_{2.5}$ sampling was conducted, concurrently, every six days at eleven sites in Israel, Jordan and the Palestinian Authority (PA)

between January and December, 2007. The sites, four in Israel, three in the Palestinian Authority and four in Jordan, were located away from specific point sources and were selected to be broadly representative of concentrations and composition for a selected site (Fig. 1). The sampling sites were located (from west to east) in: Tel Aviv, Haifa, Hebron, West Jerusalem, East Jerusalem, Nablus, Amman, Rahma, Eilat, Aqaba and Zarqa. Detailed geophysical information about the sampling sites is included as [Supplementary Data](#).

$PM_{2.5}$ mass was sampled for 24-h using a specially designed, four-channel manifold sampler (URG-3000ABC) fitted with two $PM_{2.5}$ cyclones, each sampling at flow rate of 16.7 LPM. The sampler collected $PM_{2.5}$ onto two Teflon and one quartz filter, with a bypass system in the fourth channel that allowed both cyclones to operate at their designed flow rates and each manifold to operate at 8.4 LPM. Flow rates were controlled by critical orifices located upstream of a vacuum pump and were measured at the start and end of each sampling period using calibrated rotameters.

Field technicians at each of the eleven sampling sites were carefully trained in filter handling and storage. After each sampling period, filter samples were sealed and stored in sealed Petri dishes under freezing condition to minimize volatile species loss. Samples were transported to the project's central depot at the Hebrew University in West Jerusalem, as well as to the Weizmann Institute in Rehovot, for gravimetric mass concentration measurements, and Elemental Carbon/Organic Carbon (EC/OC) analyses, respectively. XRF analyses were conducted at the Desert Research Institute (DRI) (Reno, NV); IC and GCMS analyses were conducted at the University of Wisconsin-Madison (Madison, WI).

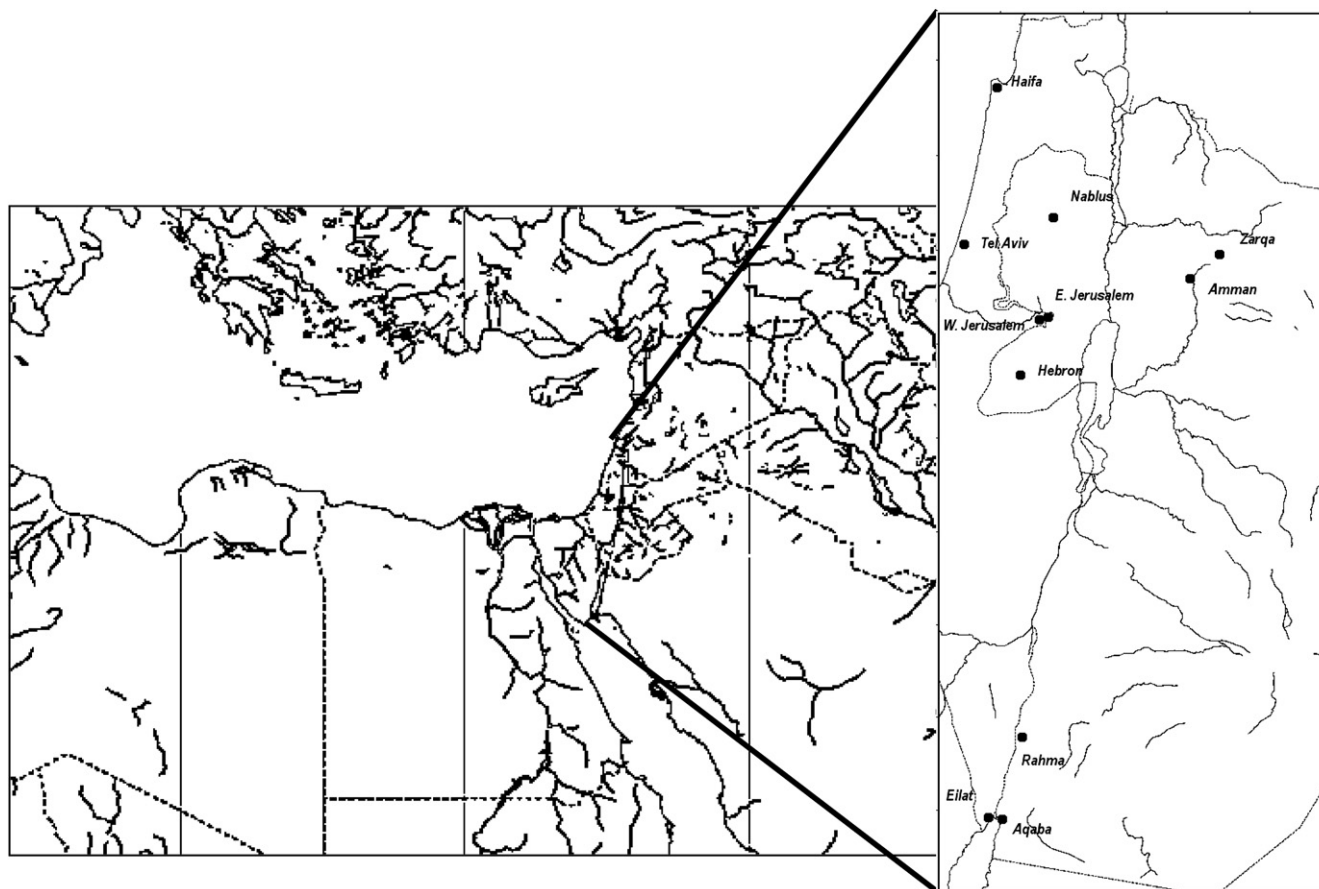


Fig. 1. Map of sites in study.

2.1. Filter analysis

Following sample collection at each of the 11 locations, the Teflon filters were analyzed for gravimetric mass concentration, major ions, and trace elements. Quartz filters were used to measure total elemental and organic carbon, as well as speciated organic carbon on a subset of filters. Gravimetric analysis was conducted on the Teflon filters before and after sampling at a specially-designated weigh room located at the Hebrew University, Jerusalem (ME5-F Microbalance, Sartorius Mechatronics) following the guidelines of the EPA's Standard Operating Procedure for PM_{2.5} Gravimetric Analysis on temperature and humidity control (EPA, 2003). In addition, inter-comparison validation measurements performed on a subset of samples ensured data quality objectives for accuracy.

Major ion concentrations were determined using ion chromatography (Dionex), comparing sampled concentrations with laboratory standards prepared from two different commercial sources of standards. One source of standards was used to calibrate the instrument, and the second source was used as check standards to confirm the calibration and quantification integrity. Trace elements analysis was conducted by the DRI using X-Ray Fluorescence (XRF) analysis. The quartz filters were analyzed using a carbon analyzer (Sunset Laboratory) to characterize total elemental carbon (EC) and organic carbon (OC) using the established NIOSH protocol (Birch and Cary, 1996).

To track any contamination due to handling, 12 sets of field blanks (one per sampling month) were collected at each site. The blank filters were stored, transported, and analyzed alongside the 24-h samples. All measurements were blank corrected using the results of the analysis of the blank filters. Field blank values were not statistically different from zero for total PM_{2.5} mass. Average masses of the blanks as a fraction of measured non-blank masses varied by component, ranging from 0.2 to 7.8% for EC and OC, respectively.

2.2. Data analysis

For this analysis, PM_{2.5} mass, as well as fine particulate SO₄²⁻, NO₃⁻, EC, OC, and crustal components were compared across sites. The 'crustal' component was calculated by aggregating the mass of the common crustal oxides of silicon, aluminum, calcium, and iron at these sites.

The spatial and temporal characteristics of the measured PM_{2.5} and its chemical components were examined using summary statistics, correlation analysis, and coefficient of variation and divergence analyses. Spearman's correlation coefficients (r_s) were calculated as a measure of the non-parametric linear association between sites or specific pollutant distributions at a given site. Strong intersite correlations are one indicator of spatial uniformity over a temporal scale, however, substantial differences in absolute concentration or in PM_{2.5} composition may still exist (Pinto et al., 2004). Coefficients of variation (CV) among the measured pollutant distributions for a given sampling day were, therefore, calculated as an additional means of assessing spatiotemporal trends in the absolute pollutant concentrations. CV was averaged over the sampling year ('mean annual CV') and defined as the mean of:

$$\frac{\sigma_i}{\mu_i}$$

where ' μ_i ' is the mean concentration of PM_{2.5} or its component across the sites on a given sampling day and ' σ_i ' is the standard deviation of these site-specific concentrations.

Finally, we estimated pairwise Coefficients of Divergence (COD) to assess the degree of uniformity in the PM_{2.5} concentrations as well as absolute differences in the PM_{2.5} chemical compositions

among the sites. COD has been used as a complementary measure to correlation analysis, to characterize spatial patterns of particulate matter in several multi-site comparative analyses (Wongphatarakul et al., 1998; Pinto et al., 2004; Kim et al., 2005; Krudysz et al., 2008). In this context, two sites may exhibit strong linear associations with each other in total PM_{2.5} mass, yet have absolute levels that differ substantially; yielding both high correlations and high COD values. For this analysis, COD was defined as:

$$COD_{jk} = \sqrt{\frac{1}{n} \sum_{i=1}^n \left(\frac{x_{ij} - x_{ik}}{x_{ij} + x_{ik}} \right)^2}$$

where ' n ' is the number of PM_{2.5} components (i.e., six); ' x_{ij} ' is the average concentration of component ' i ', measured at location ' j '; ' j ' and ' k ' represent two monitoring locations. COD approaches zero when the average PM_{2.5} measured at two sites is uniform and approaches one when PM_{2.5} concentrations and composition are dissimilar. Krudysz et al. (2008), defined CODs for pollutant distributions greater than 0.20 as being "relatively heterogeneous" or dissimilar using results from numerous sites. For the current analysis, the absolute concentrations and composition of the PM_{2.5} from each of the sites was assessed using CODs.

Since a primary objective of this study was to compare correlations of PM_{2.5} and its major components across sites, summary statistics as well as the other analyses were conducted using data collected on days when results from all sites were available for a given pollutant species ('matched data'). Dust storms occurred twice during sampling (5/30/07 and 10/09/07) leading to extreme levels of total PM_{2.5} and its crustal components throughout the entire region. Also, sampling was conducted on the Jewish holiday Lag B'Omer (5/06/07), which is celebrated in Israel with bonfires, resulting in high regional concentrations of PM_{2.5} and its biomass burning associated components. Pollutant concentrations on these sampling days are presented to anecdotally describe the potential impact of these events, but were removed from the comparative analyses since they provide little relevant information concerning typical and commonly observed spatiotemporal PM_{2.5} relationships in this region. Missing data for the chemical components were largely due to instrument error or filter contamination and were determined to be missing-at-random. Data were processed and analyzed using the SAS system, version 9.2 (SAS Institute, Cary, NC).

2.3. Factor analysis

Factor analysis is a quantitative method that searches for shared variance between different variables, here called 'factors'. Factor analysis was conducted on the total PM_{2.5} mass as an initial data reduction tool at all 11 sites. At a subset of 7 sites, factor analysis was conducted on the PM_{2.5} components as well to provide an indication of local and regional contributions to the component masses. For our analyses, the identification of a factor explaining a significant fraction of shared variance among the sites, provided a rough indication of either common source contributions or meteorologic conditions that collectively impact spatiotemporal pollutant distributions. Factor analysis was conducted using Matlab (version 7.8), with maximum likelihood as the extraction method and varimax rotation. Significant factors were defined as all factors with eigenvalues >1.0.

2.4. Trajectory analysis

Trajectory analysis was conducted for a subset of the sites during the 2007 calendar year using the HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPPLIT) model. In this analysis, HYSPPLIT

simulations used the Global Data Assimilation System (GDAS) model output generated by National Centers for Environmental Prediction (NCEP) at one-degree spatial resolution and 6-h frequency as meteorological inputs. The objective of this analysis was to provide a broad indication of the potential for trans-boundary impacts and intersite correlations during this sampling period. Given the relatively coarse spatial resolution of the meteorological inputs results from the trajectory analysis should be interpreted as being suggestive.

3. Results

Factor analysis (FA) conducted on the total $\text{PM}_{2.5}$ mass concentrations showed that 49.5% of variability in total $\text{PM}_{2.5}$ for 7 of the 11 sites was explained by a single, shared factor (Fig. 2) (Eigenvalue = 5.45; explained variability = 49.5%). A separate factor explained significant variability in total $\text{PM}_{2.5}$ mass for 3 of the remaining 4 sites (Eigenvalue = 1.65; explained variability = 15%). These three sites, Rahma, Aqaba, and Eilat, were located in close proximity (<50 km) to each other and away from the other sampling locations (Fig. 1). The fourth non-collinear site, Zarqa, was also unique in the amount of local, heavy industrial sources (Hamdi et al., 2008) (Supplementary Data). Based on these initial FA results for total $\text{PM}_{2.5}$, detailed correlation analyses on pollutant concentrations and associations among the $\text{PM}_{2.5}$ components were conducted for the seven collinear sites only.

3.1. Pollutant temporal trends and between-site correlations

For six of the seven sites, $\text{PM}_{2.5}$ mass concentrations were highest between April and August and exhibited annual minima in December or January (Fig. 3). Mean $\text{PM}_{2.5}$ concentrations over the sampling year varied considerably by site, averaging $25.7 \mu\text{g m}^{-3}$ across the sites and ranging from 19.8 to $34.9 \mu\text{g m}^{-3}$ in Haifa and Amman, respectively (Table 1). Intersite correlations for total $\text{PM}_{2.5}$ were typically moderate to strong (r_s range = 0.35–0.83; mean annual intersite $r_s = 0.66$, Table 2). Of the 21 intersite correlations for $\text{PM}_{2.5}$, only the Haifa–Amman pairwise correlation was lower than 0.50 ($r = 0.35$) (Table 2). Temporal correlation among the sites

is also evident in the monthly time series of $\text{PM}_{2.5}$ mass concentrations (Fig. 3).

Among the main $\text{PM}_{2.5}$ components, intersite correlations were strongest and concentrations most similar for SO_4^{2-} . Correlations ranged from 0.47 (East Jerusalem–Amman) to 0.92 (Tel Aviv–Haifa), with a mean intersite correlation among all sites of 0.74. SO_4^{2-} also exhibited the least amount of variability in absolute levels (mean annual CV = 28%) (Fig. 3). As expected, SO_4^{2-} concentrations peaked between May and September at all sites, reflecting enhanced secondary photochemical activity and SO_4^{2-} formation during the months of greatest sunlight intensity throughout the region and the least amount of rainfall. Results from factor analysis, conducted on the seven city subset, estimated that 74% of the SO_4^{2-} mass was shared among the sites, attributable to regional loadings. Excluding Amman, 84% of SO_4^{2-} mass was explained by a single, regional factor.

Crustal $\text{PM}_{2.5}$ concentrations followed a similar monthly pattern during the sampling year at all of the sites (Fig. 3). Correspondingly, intersite correlations for the crustal $\text{PM}_{2.5}$ component were moderate to strong (r_s range = 0.33–0.86; mean annual intersite $r_s = 0.63$) (Table 2). The mean annual CV for the crustal component, 73%, indicates heterogeneity in absolute crustal levels among the sites (Fig. 3). This value was driven largely by crustal $\text{PM}_{2.5}$ concentrations in Amman, which were substantially higher compared the other sites. The mean annual CV for crustal $\text{PM}_{2.5}$ among the six sites excluding Amman was 39%, which was lower than all the other measured pollutants except total $\text{PM}_{2.5}$ and SO_4^{2-} . Factor analysis results indicated that 56% of the crustal mass was explained by a single shared factor (i.e., meteorology or regional dust sources) among the seven sites.

There was slightly greater heterogeneity among sites for NO_3^- and OC $\text{PM}_{2.5}$ fractions compared to SO_4^{2-} and crustal mass. Intersite correlations were moderate (mean annual $r_s \approx 0.55$), with mean annual CVs of 42–45% for each of these components (Table 2, Fig. 3). Several sites exhibited weak pairwise correlations due to unique pollutant distributions of either NO_3^- or OC. Particulate NO_3^- levels in Haifa were generally not well correlated with corresponding measurements at the other sites. Between February and March, substantially elevated NO_3^- levels were measured in Haifa, but not at any of the other sites which explains weaker observed correlations between many of the other sites and Haifa. Particulate NO_3^- – SO_4^{2-} partitioning is a complex function of temperature, humidity, ammonia availability and relative pollutant concentrations at the sites and is beyond the scope of this analysis. Idiosyncratic NO_3^- peaks in Haifa, however, may also be due to the role of inorganic sea salt which has been shown to drive $\text{NO}_3^-/\text{SO}_4^{2-}$ chemistry in some locations (Song and Carmichael, 2001).

Of the key $\text{PM}_{2.5}$ components, correlations among the sites were typically weakest for EC, with a mean between-site correlation of 0.36. Stronger correlations existed for EC among the larger metropolitan areas in the study, West Jerusalem, Tel Aviv and Amman. The correlation coefficient for EC between Tel Aviv and W. Jerusalem, for example, was 0.80, compared to Tel Aviv–Nablus and Tel Aviv–E. Jerusalem correlations of 0.26 and 0.35, respectively (Table 2). On average, factor analysis results estimated that approximately half (49%) of the EC mass was attributable to local, non-shared factors, which was the relative contribution among the components measured. Analysis results including only West Jerusalem, Tel Aviv and Amman, however, identified a shared factor that explained 78.1% of the EC variability in these cities.

Within site correlations between total $\text{PM}_{2.5}$ and its individual components were also examined (Fig. 4) to provide an indication of how changes in $\text{PM}_{2.5}$ mass may reflect corresponding changes in its constituent chemical species. At all of the sites, total $\text{PM}_{2.5}$ was strongly correlated with its SO_4^{2-} component. Weaker correlations

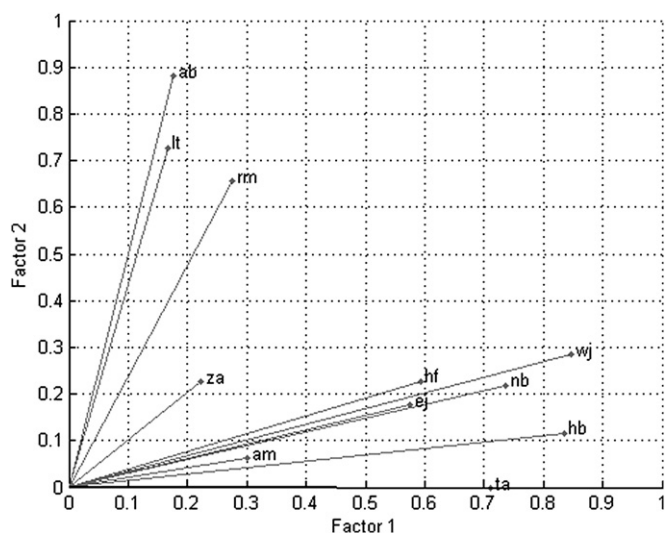


Fig. 2. Factor analysis results conducted on total $\text{PM}_{2.5}$ concentrations for all eleven sites in the study. ('ab' = Aqaba; 'am' = Amman; 'ej' = East Jerusalem; 'hb' = Hebron; 'hf' = Haifa; 'lt' = Eilat; 'nb' = Nablus; 'rm' = Rahma; 'ta' = Tel Aviv; 'wj' = West Jerusalem; 'za' = Zarqa.)

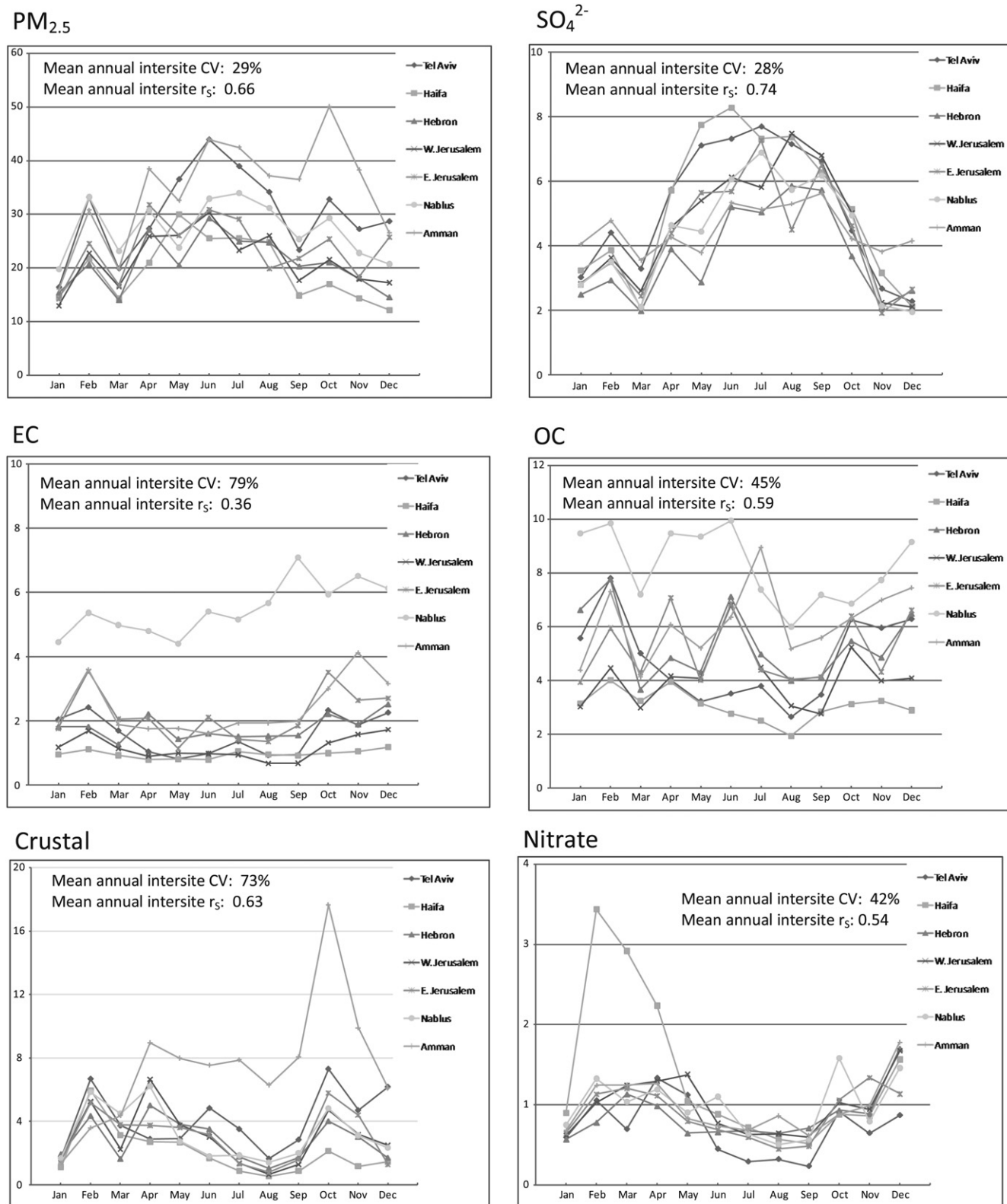


Fig. 3. Monthly time series of PM_{2.5} and PM_{2.5} components. Crustal component includes aggregation of oxides of silicon, aluminum, calcium, and iron.

were found between total PM_{2.5} and crustal and organic carbon, with a particularly strong PM_{2.5}-crustal correlation observed in Amman ($r_s = 0.88$). In contrast, there were generally weak correlations between PM_{2.5} and its corresponding EC and NO₃ fractions at any of the sites.

3.2. Coefficients of divergence

Pairwise COD values were plotted against correlation coefficients for total PM_{2.5} to provide an indication of how well linear association agrees with absolute PM_{2.5} levels and composition

Table 1
Summary of PM_{2.5} mass concentrations and PM_{2.5} chemical components across the seven sites. All concentrations are reported in $\mu\text{g m}^{-3}$. Crustal component includes aggregation of oxides of silicon, aluminum, calcium, and iron.

| | Total fine particulate matter | | | | | Sulfate | | | | | Elemental carbon | | | | |
|--------------|-------------------------------|------|------|--------|------|---------|------|-----|--------|------|------------------|------|------|--------|------|
| | N | Mean | SD | Median | Max | N | Mean | SD | Median | Max | N | Mean | SD | Median | Max |
| Tel Aviv | 43 | 30.7 | 11.2 | 31.2 | 70.8 | 50 | 5.2 | 3.1 | 4.4 | 13.6 | 52 | 1.5 | 0.9 | 1.3 | 3.7 |
| Haifa | 43 | 19.8 | 7.7 | 20.1 | 36.8 | 50 | 5.2 | 3.1 | 4.6 | 13.4 | 52 | 1.0 | 0.3 | 0.9 | 1.7 |
| Hebron | 43 | 21.1 | 7.5 | 20.1 | 37.7 | 50 | 3.8 | 2.0 | 3.4 | 9.0 | 52 | 1.8 | 0.8 | 1.5 | 4.4 |
| W. Jerusalem | 43 | 21.7 | 7.5 | 20.9 | 36.9 | 50 | 4.6 | 2.6 | 4.3 | 11.2 | 52 | 1.1 | 0.6 | 0.9 | 3.4 |
| E. Jerusalem | 43 | 23.8 | 7.9 | 24.1 | 38.1 | 50 | 4.4 | 2.4 | 4.1 | 10.7 | 52 | 2.2 | 1.3 | 1.7 | 6.6 |
| Nablus | 43 | 27.6 | 7.9 | 26.7 | 45.9 | 50 | 4.3 | 2.6 | 3.8 | 11.0 | 52 | 5.6 | 1.9 | 5.4 | 9.0 |
| Amman | 43 | 34.9 | 15.1 | 29.9 | 72.2 | 50 | 4.5 | 1.9 | 4.5 | 9.6 | 52 | 2.4 | 1.5 | 1.9 | 7.5 |
| | | | | | | | | | | | | | | | |
| | Organic carbon | | | | | Crustal | | | | | Nitrate | | | | |
| | N | Mean | SD | Median | Max | N | Mean | SD | Median | Max | N | Mean | SD | Median | Max |
| Tel Aviv | 52 | 4.7 | 2.5 | 4.1 | 12.7 | 50 | 4.0 | 3.7 | 2.6 | 16.2 | 46 | 0.70 | 0.57 | 0.57 | 3.09 |
| Haifa | 52 | 3.0 | 1.3 | 2.7 | 6.4 | 50 | 1.9 | 2.7 | 0.8 | 13.7 | 46 | 1.36 | 1.56 | 0.91 | 8.14 |
| Hebron | 52 | 5.3 | 2.5 | 4.7 | 13.2 | 50 | 2.7 | 2.7 | 1.7 | 11.0 | 46 | 0.91 | 0.52 | 0.77 | 3.20 |
| W. Jerusalem | 52 | 4.1 | 2.4 | 3.8 | 15.7 | 50 | 2.9 | 3.5 | 1.6 | 17.9 | 46 | 1.02 | 0.57 | 0.87 | 2.86 |
| E. Jerusalem | 52 | 5.2 | 2.8 | 4.7 | 14.9 | 50 | 2.9 | 3.0 | 1.7 | 13.1 | 46 | 0.92 | 0.60 | 0.79 | 3.58 |
| Nablus | 52 | 8.2 | 2.9 | 8.0 | 15.9 | 50 | 3.1 | 3.9 | 1.9 | 22.5 | 46 | 1.01 | 0.64 | 0.81 | 3.73 |
| Amman | 52 | 6.2 | 3.5 | 5.4 | 18.9 | 50 | 7.7 | 6.1 | 6.3 | 28.0 | 46 | 1.02 | 0.53 | 0.80 | 2.60 |

(Fig. 5). Sites highly correlated with each other having low CODs (i.e., points in the lower right quadrant of Fig. 5), indicate similar temporal variation as well as the absolute concentrations and composition of PM_{2.5}. Nablus, in particular, had PM_{2.5} levels and composition that was dissimilar to the other sites. CODs between Nablus and the other six sites were all above 0.20, which has been used previously to mark pollutant heterogeneity (Pinto et al., 2004). These high CODs were largely due to higher concentrations of both EC and OC in Nablus compared to the other sites. Amman also was shown to have PM_{2.5} levels and composition that were generally dissimilar to the others sites, with the COD between Amman and E. Jerusalem being the only value less than 0.20. In contrast, W. Jerusalem was shown to have PM_{2.5} that was fairly similar in concentration and composition to many of the other sites. Pairwise CODs for W. Jerusalem with E. Jerusalem, Tel Aviv, Hebron and Haifa, respectively, were all less than 0.15 denoting relative homogeneity among the sites with W. Jerusalem.

3.3. Trajectory analysis

Trajectory analysis was conducted daily for the entire sampling year and arbitrarily aggregated by calendar quarter to identify broad seasonal patterns in air mass movement that may affect between-site correlation patterns. Maps showing 48-h backward trajectories from West Jerusalem are presented as representative for the seven sites. West Jerusalem was selected as a background site, since it is centrally-located within the sampling domain and it typically was the site most strongly correlated with other sites in the study. Results showed a clear dominance of slowly moving, westerly trajectories originating in central Europe and N. Africa and limited vertical atmospheric mixing indicative of highly stable conditions throughout the entire year (Fig. 6). Back trajectory analyses conducted for Amman were noticeably different from that observed for West and East Jerusalem, Hebron and Tel Aviv (Supplementary Data), indicating the presence of north- and southeasterly trajectories occurring for much of the year. Many of the low, ground-level trajectories were slow, suggesting poor vertical dilution and greater potential for ground-level impact from sources originating east of Amman.

Three extreme PM events coincided with our sampling schedule and resulted in elevated total PM_{2.5} concentrations throughout all the study sites. Mean PM_{2.5} concentrations during the two dust storms on May 30th and October 9th were 110.2 and 56.3 $\mu\text{g m}^{-3}$,

respectively. Not surprisingly, crustal PM concentrations were also elevated during the dust storms, with cross-site concentrations averaging 47.3 and 15.5 $\mu\text{g m}^{-3}$ for this component for the May and October events, respectively. EC and OC levels were not elevated during these events. Pollutant measurements on Jewish holiday of Lag B'Omer (May 6th) were characterized by high total PM_{2.5} (mean cross-site concentrations = 68.9 $\mu\text{g m}^{-3}$), OC (15.0 $\mu\text{g m}^{-3}$), EC (4.1 $\mu\text{g m}^{-3}$) and crustal (20.1 $\mu\text{g m}^{-3}$) concentrations. Elevated pollutant levels were observed throughout all the sites in our study area, indicating the regional impact of these events.

4. Discussion

This project constituted the first investigation of spatiotemporal correlation of PM_{2.5} and its major components in Israeli, Jordanian and Palestinian sites and the first time chemically-resolved ambient PM_{2.5} components were measured in Jordan and Palestine. Annual PM_{2.5} levels varied by site, however, mean concentrations from all sites exceeded accepted international air quality standards for annual PM_{2.5} mass (e.g., World Health Organization = 10 $\mu\text{g m}^{-3}$; U.S. Environmental Protection Agency = 15 $\mu\text{g m}^{-3}$). Mean PM_{2.5} in Tel Aviv and Amman were the highest among the seven locations examined, with levels that were 2–3 times international standards.

The heterogeneity in mean concentrations, from 19.8 $\mu\text{g m}^{-3}$ in Haifa to 34.9 $\mu\text{g m}^{-3}$ in Amman, was substantial but consistent with pollutant ranges from in other published PM_{2.5} monitoring results from the region. To assess the plausibility of our observed range of mean PM_{2.5} concentrations across the sites, we examined data posted by the Israeli Ministry of Environment (MOE) from a network of 20 continuous PM_{2.5} monitoring sites throughout the country during 2007 (www.sviva.gov.il). While the averaging times of these monitors with those used in our study differ (i.e., continuous vs. 24 h integrated), the results from the government network were comparable to our findings, exhibiting an annual cross-site mean PM_{2.5} concentration of 23 $\mu\text{g m}^{-3}$ (compared to 24 $\mu\text{g m}^{-3}$ for our sites) and a range of 18–28 $\mu\text{g m}^{-3}$ among the individual monitoring sites.

PM_{2.5} concentrations measured at our Tel Aviv site were considerably higher than those measured at four other MOE sites in Tel Aviv (annual PM_{2.5} concentrations of 25, 24, 22, 20 $\mu\text{g m}^{-3}$), as well as our other coastal site in Haifa. The relative composition of the PM at this site, especially the OC and EC, suggests that this high mean value is not mobile source related. Differences between our

Table 2

Pairwise Spearman's correlation coefficients (r_s) by component. Crustal component includes aggregation of oxides of silicon, aluminum, calcium, and iron. 'Mean' represents mean intersite r_s for a given sampling location.

| | Tel Aviv | Haifa | Hebron | W. Jerusalem | E. Jerusalem | Nablus | Amman |
|-------------------------|----------|-------|--------|--------------|--------------|--------|-------|
| PM_{2.5} | | | | | | | |
| Tel Aviv | | 0.71 | 0.64 | 0.71 | 0.63 | 0.60 | 0.53 |
| Haifa | 0.71 | | 0.57 | 0.73 | 0.50 | 0.60 | 0.35 |
| Hebron | 0.64 | 0.57 | | 0.83 | 0.70 | 0.78 | 0.64 |
| W. Jerusalem | 0.71 | 0.73 | 0.83 | | 0.77 | 0.66 | 0.61 |
| E. Jerusalem | 0.63 | 0.50 | 0.70 | 0.77 | | 0.62 | 0.61 |
| Nablus | 0.60 | 0.60 | 0.78 | 0.66 | 0.62 | | 0.57 |
| Amman | 0.53 | 0.35 | 0.64 | 0.61 | 0.61 | 0.57 | |
| Mean | 0.64 | 0.58 | 0.69 | 0.72 | 0.64 | 0.64 | 0.55 |
| EC | | | | | | | |
| Tel Aviv | | 0.54 | 0.35 | 0.80 | 0.35 | 0.26 | 0.66 |
| Haifa | 0.54 | | 0.34 | 0.47 | 0.40 | 0.17 | 0.40 |
| Hebron | 0.35 | 0.34 | | 0.44 | 0.28 | 0.38 | 0.35 |
| W. Jerusalem | 0.80 | 0.47 | 0.44 | | 0.40 | 0.21 | 0.73 |
| E. Jerusalem | 0.35 | 0.40 | 0.28 | 0.40 | | 0.27 | 0.28 |
| Nablus | 0.26 | 0.17 | 0.38 | 0.21 | 0.27 | | 0.47 |
| Amman | 0.66 | 0.40 | 0.35 | 0.73 | 0.28 | 0.47 | |
| Mean | 0.49 | 0.39 | 0.36 | 0.51 | 0.33 | 0.30 | 0.48 |
| Crustal | | | | | | | |
| Tel Aviv | | 0.70 | 0.56 | 0.73 | 0.50 | 0.60 | 0.56 |
| Haifa | 0.70 | | 0.63 | 0.75 | 0.61 | 0.78 | 0.33 |
| Hebron | 0.56 | 0.63 | | 0.86 | 0.61 | 0.68 | 0.62 |
| W. Jerusalem | 0.73 | 0.75 | 0.86 | | 0.76 | 0.82 | 0.62 |
| E. Jerusalem | 0.50 | 0.61 | 0.61 | 0.76 | | 0.66 | 0.44 |
| Nablus | 0.60 | 0.78 | 0.68 | 0.82 | 0.66 | | 0.49 |
| Amman | 0.56 | 0.33 | 0.62 | 0.62 | 0.44 | 0.49 | |
| Mean | 0.61 | 0.63 | 0.66 | 0.76 | 0.60 | 0.67 | 0.51 |
| Sulfate | | | | | | | |
| Tel Aviv | | 0.92 | 0.67 | 0.84 | 0.69 | 0.86 | 0.56 |
| Haifa | 0.92 | | 0.69 | 0.84 | 0.72 | 0.84 | 0.50 |
| Hebron | 0.67 | 0.69 | | 0.89 | 0.72 | 0.83 | 0.61 |
| W. Jerusalem | 0.84 | 0.84 | 0.89 | | 0.77 | 0.90 | 0.62 |
| E. Jerusalem | 0.69 | 0.72 | 0.72 | 0.77 | | 0.73 | 0.47 |
| Nablus | 0.86 | 0.84 | 0.83 | 0.90 | 0.73 | | 0.60 |
| Amman | 0.56 | 0.50 | 0.61 | 0.62 | 0.47 | 0.60 | |
| Mean | 0.76 | 0.75 | 0.73 | 0.81 | 0.68 | 0.79 | 0.56 |
| OC | | | | | | | |
| Tel Aviv | | 0.59 | 0.72 | 0.74 | 0.44 | 0.58 | 0.68 |
| Haifa | 0.59 | | 0.53 | 0.51 | 0.42 | 0.62 | 0.55 |
| Hebron | 0.72 | 0.53 | | 0.76 | 0.56 | 0.71 | 0.70 |
| W. Jerusalem | 0.74 | 0.51 | 0.76 | | 0.52 | 0.62 | 0.81 |
| E. Jerusalem | 0.44 | 0.42 | 0.56 | 0.52 | | 0.56 | 0.45 |
| Nablus | 0.58 | 0.62 | 0.71 | 0.62 | 0.56 | | 0.64 |
| Amman | 0.68 | 0.55 | 0.70 | 0.81 | 0.45 | 0.64 | |
| Mean | 0.63 | 0.54 | 0.66 | 0.66 | 0.49 | 0.62 | 0.64 |
| Nitrate | | | | | | | |
| Tel Aviv | | 0.71 | 0.44 | 0.54 | 0.55 | 0.59 | 0.45 |
| Haifa | 0.71 | | 0.35 | 0.43 | 0.58 | 0.57 | 0.44 |
| Hebron | 0.44 | 0.35 | | 0.77 | 0.70 | 0.64 | 0.68 |
| W. Jerusalem | 0.54 | 0.43 | 0.77 | | 0.69 | 0.62 | 0.69 |
| E. Jerusalem | 0.55 | 0.58 | 0.70 | 0.69 | | 0.66 | 0.68 |
| Nablus | 0.59 | 0.57 | 0.64 | 0.62 | 0.66 | | 0.74 |
| Amman | 0.45 | 0.44 | 0.68 | 0.69 | 0.68 | 0.74 | |
| Mean | 0.54 | 0.51 | 0.60 | 0.62 | 0.64 | 0.64 | 0.61 |

filter-based and MOE's continuous PM sampling methods may explain the magnitude of this discrepancy and have resulted in a sampling artifact, related to the treatment of particle-bound water. Thus, a more complete volatilization of this fraction may have occurred using the continuous method compared to the filter methods. Similarly, it is possible that the differences in PM hygroscopicity are also a factor associated with the pronounced

differences between the annual PM_{2.5} concentrations in Tel Aviv and Haifa. Although the SO₄²⁻ ion, sodium ion and ammonium ion concentrations in the two coastal cities of Tel Aviv and Haifa were very similar, chloride levels in Haifa were about 36% of the levels in Tel Aviv and nitrate levels in Haifa were about 55% of the levels in Tel Aviv. This suggests that the aerosol in Tel Aviv may be less neutralized than the aerosol in Haifa (Seinfeld and Pandis, 1998), which will increase the affinity of the aerosol in Tel Aviv for water as compared to the aerosol in Haifa.

Despite these prominent examples of difference in total PM_{2.5} concentrations, the findings clearly indicated that total PM_{2.5} mass was relatively homogenous among many of the sites as shown from strong between-site correlations and low daily CVs. For total PM_{2.5} mass, strong spatiotemporal correlations likely reflected the similar contribution from regional PM sources at these sites, as well as synoptic meteorological conditions which have been shown to affect atmospheric mixing and dispersion processes regionally (Dayan, 1986; Weinroth et al., 2006; Asaf et al., 2008). Results from our back trajectory analysis support this interpretation in showing a clear predominance of regional, slow-moving, westerly air movement for much of the year, originating in central Europe and North Africa.

Our results also suggest that the similarity of total mass was largely driven by SO₄²⁻ and crustal PM_{2.5} components. This was not unexpected for SO₄²⁻ (Luria et al., 1986; Suh et al., 1995; Burton et al., 1996), which is formed photochemically through secondary atmospheric processes. Previous studies have suggested that coal fired power plant emissions originating in southeastern Europe are a main contributor to SO₄²⁻ precursors in sites throughout Israel (Luria et al., 1996; Asaf et al., 2008). SO₄²⁻ in Amman provided an interesting exception to the uniformity across the study area, with concentrations that were similar in absolute levels, yet weakly correlated with each of the other sites. This finding may reflect the impact of local primary emissions from sulfur-rich diesel and the numerous industrial processing sites in Zarqa, located 15 km to the northeast of Amman. These sites are potential sources of SO₄²⁻ chemical precursors including gaseous SO₂ (Hamdi et al., 2008), which given their geographic proximity and the results from the back trajectory analysis identifying slow-moving easterly trajectories, may lead to the formation of a photochemical pollution 'hot-spot' in this area. Alternatively, Amman may have unique sources of SO₄²⁻ compared to the other six sites, including the heavy use of construction-related gypsum (CaSO₄·H₂O) or low grade diesel, which contains up to 1.2% sulfur (Al-Momani et al., 2002, 2008).

While not as strong as SO₄²⁻, crustal PM_{2.5} was also shown to be homogeneously distributed with moderate correlations among the sampling sites. Again, Amman served as an exception to this pattern of similarity, which may be due to calcium-rich dust emitted from local construction activity (Al-Momani et al., 2008). In the current analysis, the crustal category was heavily comprised of elemental silicon, suggestive of a regional source contribution from wind-blown desert dust (Mamane et al., 2008). It is worth noting that strong correlation among the sites in the crustal component generally contrasts with results from the U.S. and elsewhere, where crustal PM_{2.5} has been shown to be among the most heterogeneously distributed PM_{2.5} components between cities and regions (Pinto et al., 2004). In many parts of the U.S., crustal PM_{2.5} is associated with locally-generated resuspended road dust, rather than wind-blown desert sources, which accounts for weak between-site correlations.

Strong between-site spatiotemporal correlations observed for the SO₄²⁻ and crustal components were not consistent for other PM_{2.5} components. Despite the close proximity of the seven sites in this analysis, there were pronounced differences among the cities for EC and, to a lesser degree, OC. EC, in particular, exhibited

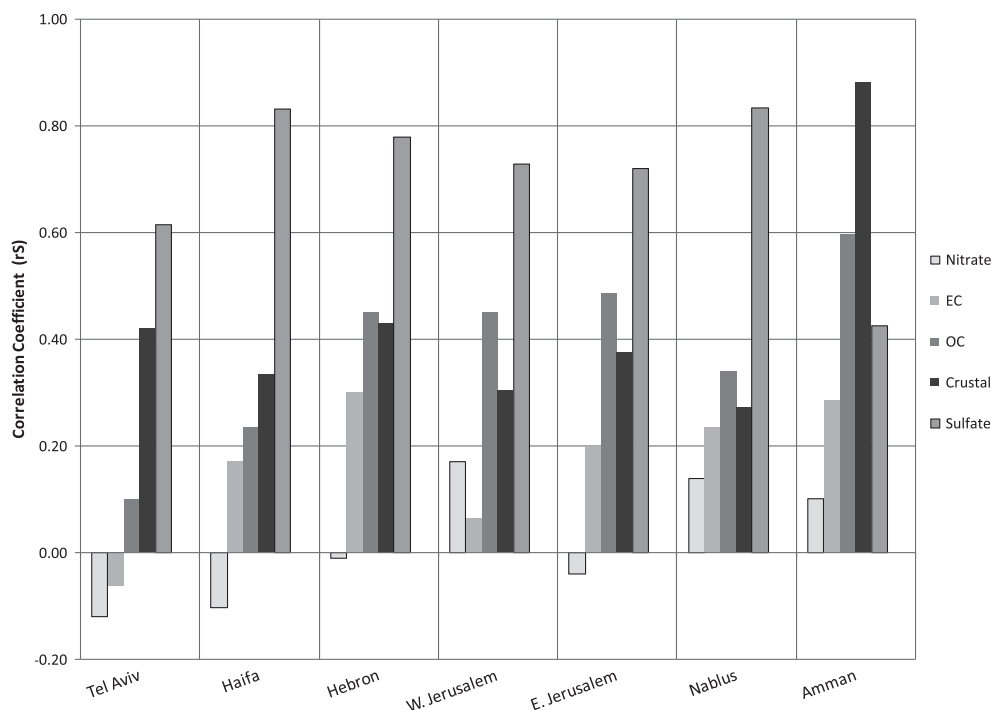


Fig. 4. Within site Spearman's correlation coefficient (r_s) between total $PM_{2.5}$ and its major components. Crustal component includes aggregation of oxides of silicon, aluminum, calcium, and iron.

spatiotemporal trends that were indicative of strong local source contributions. This finding is consistent with previous studies showing high within-city spatial heterogeneity for EC, given its local, primary source contributions (Krudysz et al., 2008). Tel Aviv and Nablus, located 50 km from each other, serve to highlight the importance of local sources and their impact on $PM_{2.5}$ composition. The COD between these closely situated sites, 0.299 (Fig. 5), was among the highest in this analysis of any two sites (i.e., demonstrating dissimilarity), despite the moderate Tel Aviv–Nablus correlation for total $PM_{2.5}$ mass. Much of this dissimilarity was due

to discrepancies in the measured EC concentrations between the two cities. Similarly, the observed COD between Haifa and Tel Aviv was high (COD = 0.232) (Fig. 5), indicative of moderate heterogeneity, despite relatively strong observed intersite correlations for total $PM_{2.5}$ ($r_s = 0.73$). Concentrations of $PM_{2.5}$ and its components were higher in Tel Aviv than Haifa, with the exception of particulate nitrate, which was almost twice as high in Haifa as compared to Tel Aviv.

Interestingly, there were the moderate to strong EC correlations ($r > 0.65$) among the large metropolitan cities, West Jerusalem, Tel Aviv and Amman. For these three cities, a single, shared factor explained approximately 80% of the variability in EC. Correlations among the large cities existed despite considerable differences in absolute EC concentration, as well as geographic distance. It is possible that this finding is attributable to typical diesel emission profiles occurring in these large urban areas. For these larger cities (i.e., West Jerusalem, Tel Aviv and Amman), EC sources from the fleet of buses and cars typical for many urban areas predominate and likely drive spatiotemporal EC distributions. The similar temporality of these emissions (i.e., high levels during weekday rush hours) combined with meteorological synoptic conditions may contribute to the strong temporal associations among the larger cities. Synoptic conditions characterized by frequent summertime subtropical high pressure are fairly typical during specific times of the year. In measurements collected over three years, Dayan and Rodnizki (1999) found that these conditions can persist in more than 25 days a month between July and September. Previous studies have also identified a 'shallow Persian trough', characterized by stagnation condition, a shallow mixing layer, and urban poor ventilation; leading to regional increases in ground-level urban air pollution levels (Ranmar et al., 2002). Smaller, local sources of EC in these large cities probably did not contribute substantially to total measured EC levels and did not impact the temporal distributions. In contrast, for the smaller cities in this analysis, such as Nablus, local non-traffic sources likely did impact

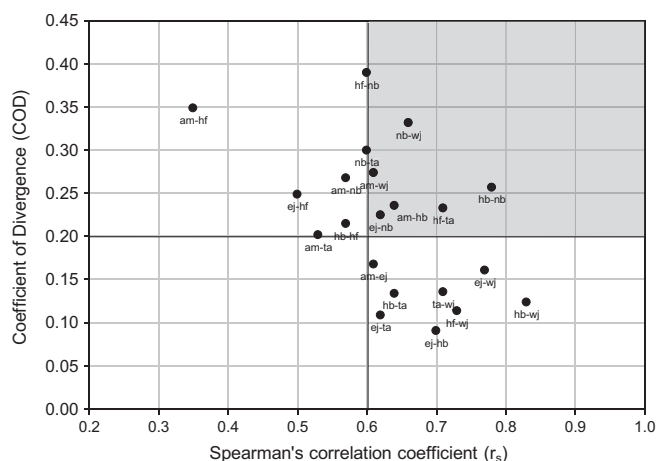


Fig. 5. Pairwise Spearman's correlation coefficients for total $PM_{2.5}$ as a predictor of corresponding pairwise Coefficients of Divergence (COD) among the seven sites. Shaded area denotes discrepant pairs with COD's > 0.200, indicative of heterogeneity in composition and/or absolute concentration, with corresponding r_s 's > 0.60 indicative of moderate to strong correlation in total $PM_{2.5}$. ('am' = Amman; 'ej' = East Jerusalem; 'hb' = Hebron; 'hf' = Haifa; 'nb' = Nablus; 'ta' = Tel Aviv; 'wj' = West Jerusalem.)

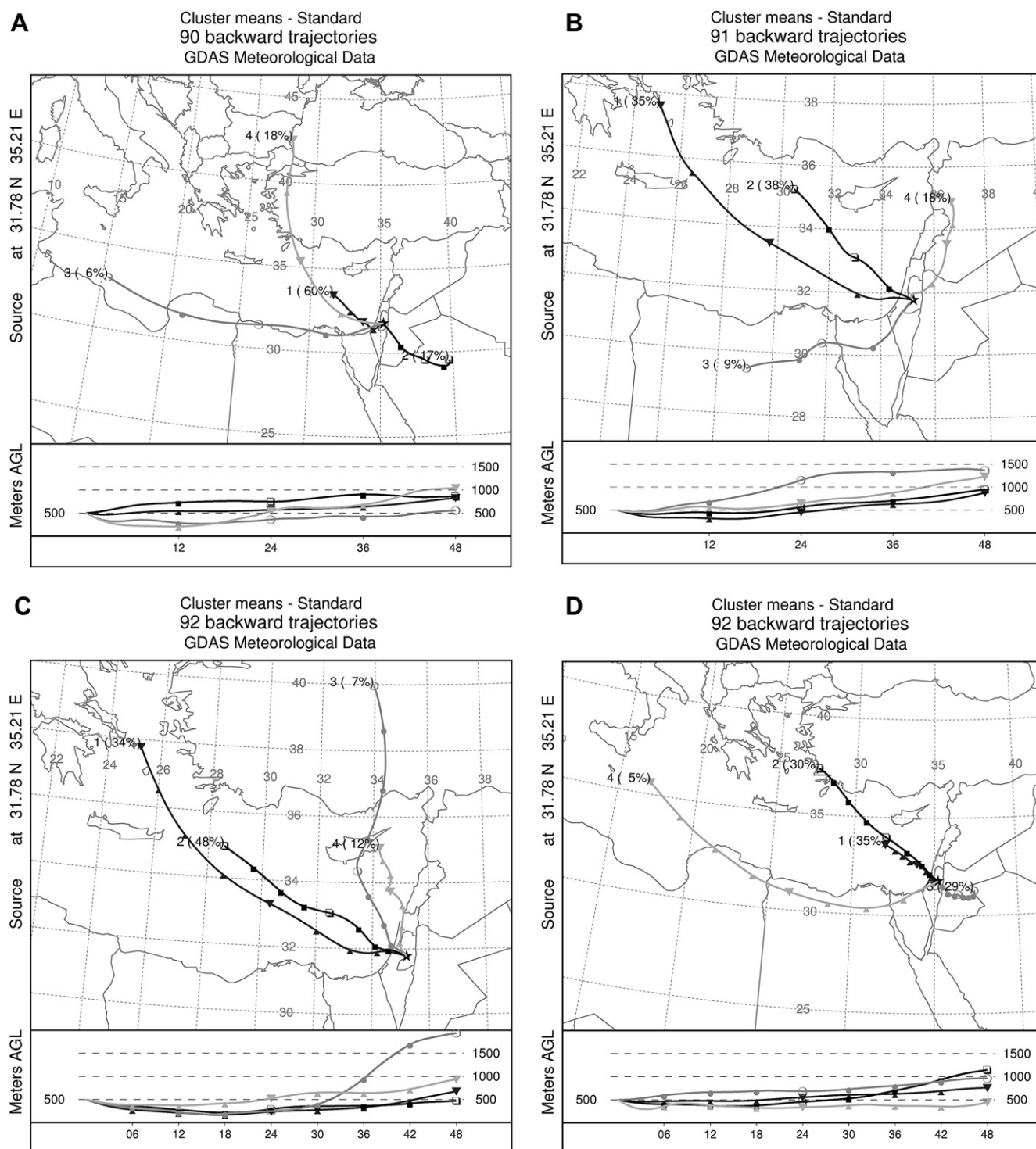


Fig. 6. Mean 48-h backward trajectories for West Jerusalem averaged by calendar quarter. A = January–March; B = April–June; C = July–September; D = October–December.

total EC levels. Nablus, for example, a prominent source of diesel emissions is four large diesel generators located throughout the small city. Temporal emission profiles different from traffic emissions, would result in EC concentrations dissimilar to those observed in the large cities.

In contrast to EC, OC has numerous regional and local sources and this likely led to a mixed pattern of correlation among the sampling locations. Using data collected during this study, we

recently conducted source apportionment using organic fingerprinting for a subset of samples at the East and West Jerusalem sites (von Schneidemesser et al., 2009). Considerable differences existed in the particulate organic composition between these two adjacent sites for many organic species, highlighting the importance of local sources of OC. The current results and relatively strong correlations among some of the sites for total OC, however, indicate that regionally formed, secondary organic aerosol also comprises

a substantial fraction of this component. It should be noted that the association between traffic-related precursor emissions along the Israeli coast and the formation of ozone and other secondary products of atmospheric oxidation in sites in eastern Israel and western Jordan, has been previously investigated (Lifshitz et al., 1988; Ranmar et al., 2002; Weinroth et al., 2006).

5. Summary

These findings have important implications for airshed management approaches. Despite the relatively small geographic domain for the seven sites in this analysis, our findings suggest that a regional approach to reduce total PM_{2.5} may produce uneven results. The variability in PM_{2.5} mass and its chemical components for most of the sites mainly reflected corresponding variability in either its SO₄^{2−} or crustal components. In contrast, total PM_{2.5} was poorly correlated with EC and, to a lesser extent, OC, two important PM_{2.5} components linked with adverse health effects in numerous epidemiologic and toxicologic studies (Pope and Dockery, 2006). Thirteen of the 21 pairwise COD comparisons (62%) were above 0.200, providing further evidence of either dissimilar PM_{2.5} absolute concentrations or composition across the sites. As new airshed management strategies and public health interventions are implemented throughout the Middle East, our findings support regulatory strategies that target reduction in specific components and emission sources of PM_{2.5} along with total mass.

Finally, although examining PM_{2.5} trends and associations were the primary objective of this research initiative, other aspects of this project warrant specific mention. Notably, this was the first time air quality researchers from this region, worked together to investigate particle air pollution. The substantial scientific cooperation, intellectual exchange and coordinated logistical effort among the research teams necessitated by the parallel sampling design, often served as a stark juxtaposition to the frequently difficult political relationships among the various political entities of this region. Moreover, the successful completion of this project served as a meaningful, albeit rare, example of the potential for scientific discourse to serve to bridge divergent political, social and religious affiliations. Further analyses investigating the PM_{2.5} components and trends are currently ongoing and will hopefully provide greater insight into the sources and impacts of PM_{2.5} pollution and the health improvements across this region.

Appendix. Supplementary data

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.atmosenv.2010.04.007.

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